ENERGY DISTRIBUTIONS OF NEUTRONS USING THE BERYLLIUM DETECTOR METHOD

Abridged from the paper of J.R.D. Copley, D. A. Neumann and W. A. Kamitakahara:
"Energy distributions of neutrons scattered from C_{60} by the beryllium detector method"
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INTRODUCTION

In the *Proceedings of the 1960 Symposium on Inelastic Scattering of Neutrons in Solids and Liquids*, Bertram Brockhouse gives a detailed description of the Chalk River triple-axis spectrometer and discusses the principal methods of operation of such instruments.[1] In the same volume, in a paper entitled *"Energy Distributions of Neutrons Scattered from Graphite, Light and Heavy Water, Ice, Zirconium Hydride, Lithium Hydride, Sodium Hydride and Ammonium Chloride by the Beryllium Detector Method"* [2], Woods, Brockhouse, Sakamoto, and Sinclair describe experiments to measure densities of states in various materials using the recently conceived filter-analyzer technique.

EXPERIMENTAL ASPECTS

Data were obtained using a filter analyzer spectrometer similar to the instrument described by Woods *et al*.[2]. A monoenergetic neutron beam is selected from the white reactor beam using a monochromator crystal that reflects neutrons to the sample position. The energy analysis of the scattered neutrons is however performed differently, using a low-pass polycrystalline filter instead of the crystal analyzer that is used in the triple-axis instrument. Neutrons with wavelengths longer than the Bragg cutoff $2d_{\max}$ of the filter material (where $d_{\max}$ is its maximum interplanar spacing) are transmitted with no attenuation due to Bragg scattering, but shorter wavelength neutrons are strongly attenuated. Phonon processes within the filter can still scatter the long wavelength ($\lambda > 2d_{\max}$)neutrons, and it is therefore advantageous to cool the filter.

The filtering characteristics of polycrystalline beryllium and graphite are evident from the plots of their cross sections as a function of energy. If the filter material is beryllium the cutoff energy is ~5 meV, *i.e.* scattered neutrons with energies less than 5 meV are transmitted with high probability. On the other hand polycrystalline graphite only transmits neutrons with energies less than ~1.8 meV. The filter that we used in our experiment consisted of beryllium and graphite. The entire filter was cooled with liquid nitrogen to ~77

K. The filter included beryllium, in addition to the graphite which determines the cutoff energy of the filter, because Be is a somewhat more effective filter for neutrons with energies above 5 meV. It also tends to contain fewer defects that give rise to undesirable small angle scattering of long wavelength neutrons. Furthermore, there is less scattering due to low-energy phonons in Be than graphite.

In filter analyzer spectrometry, the energy transfer associated with the detected neutrons is scanned by varying the incident neutron energy E_0. The mean energy transfer $E = \hbar\omega$ is then equal to $E_0 - E'$ where E' is the mean energy of the scattered neutrons. For the beryllium/graphite combination filter, E' is ~1.2 meV, and the measured Gaussian-equivalent energy resolution is ~1.1 meV FWHM (full width at half maximum height).

The filter analyzer method has a number of advantages over other methods of measuring scattered neutron energy distributions at relatively large energy transfers. The advantages and disadvantages of the method, as described in ref.[2], are the following:

1. The efficiency of the analyzing system remains constant and therefore the observed intensity does not require a correction for the poorly known sensitivity function of the analyzer and the counter.
2. On the other hand the energy variation of the sensitivity of the thin fission counter monitor in the incident beam is well known.
3. Because an energy loss process is used, the high frequency transitions are not attenuated by the Boltzmann factor as in energy gain methods. Thus measurements at low temperature are possible.
4. The use of the beryllium-shielded detector eliminates higher order effects in the analyzing system. This also implies that the second and higher order contaminant neutrons in the incident beam are not as apt to be important as in a crystal analyzing system.
5. Since the counter is required to be sensitive to very low energy neutrons only, a low [detector gas] pressure ... may be used. The fast neutron background is thus reduced while high efficiency for counting slow neutrons is preserved.
6. The experimental conditions can be made such that the counter presents a large solid angle to the specimen and as a result the intensity is high. [The solid angle accepted by the [old] detector in the filter analyzer spectrometer at NIST is approximately $9^o \times 9^o$.]
7. Insofar as the monitor counter has a $1/v$ sensitivity function [where *v* represents neutron velocity] it eliminates the factor k'/k_0 [k' and k_0 are the wave vectors of the incident and scattered neutrons, respectively] from the expression for the inelastic differential cross-section [see eq. (1) below], thus removing all explicit dependence on the initial and final neutron energies and leaving only the Van Hove scattering function, $S(Q,\omega)$, which depends only on momentum and energy transfers ... .

Disadvantages of the method are:

1. The wave vector of the scattered neutrons, k', is small so that the range of momentum transfers, Q, is restricted for a given energy transfer ... .The experiment thus encompasses a rather narrow band through $\omega - Q$ space.
2. High energy transfers are inevitably accompanied by high momentum transfers resulting in Doppler broadening of the high-energy peaks or, in another language, multiple phonon transfer.

THE SCATTERING CROSS SECTION

The double differential cross section per unit solid angle Ω, per unit energy transfer, for one-phonon scattering in neutron energy-loss scattering by an elemental coherent scatterer such as carbon, may be written as follows [3,4]:

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k'}{k_0}\frac{(2\pi)^3}{v_0}\frac{\sigma_c}{8\pi M}\frac{Q^2}{\omega}\left[n(\omega)+1\right]\sum_{\boldsymbol{\tau}}\sum_{\mathbf{q}j}\delta\left[\omega-\omega_j(\mathbf{q})\right]\delta(\mathbf{Q}-\mathbf{q}-\boldsymbol{\tau})\left|\sum_d e^{i\mathbf{Q}\mathbf{r}_d}\left[\hat{Q}.\hat{e}_{dj}(\mathbf{q})\right]e^{-\mathrm{W}_d(\mathbf{Q})}\right|^2 \quad (1)$$

where v_0 is the volume of the unit cell, σ_c is the coherent scattering cross section per atom, M is the mass of an atom, $n(\omega)$ is the Bose factor, $\boldsymbol{\tau}$ is a reciprocal lattice vector, $\omega_j(\mathbf{q})$ is the frequency of the j th normal mode with wave vector $\mathbf{q}$, $\hat{e}_{dj}(\mathbf{q})$ is the corresponding eigenvector component for atom d, r_d is the equilibrium position of atom d, $e^{-\mathrm{W}_d(\mathbf{Q})} = e^{-Q^2\left\langle u_d^2\right\rangle/2}$ is its Debye-Waller amplitude factor ($\left\langle u_d^2\right\rangle$ being the mean square displacement of atom d), and the sum within the squared quantity is a sum over atoms in the unit cell.

For a collection of N_m isolated elemental molecules (1) may be simplified, recognizing that mode frequencies and eigenvectors no longer depend on $\mathbf{q}$. Additionally, performing the sum over $\boldsymbol{\tau}$, we obtain a result that we shall call the "single-molecule approximation".

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k'}{k_0}\frac{N_m\sigma_c}{8\pi M}\frac{Q^2}{\omega}\left[n(\omega)+1\right]\sum_j(\omega-\omega_j)\left|\sum_d e^{i\mathbf{Q}\mathbf{r}_d}\left[\hat{Q}.\hat{e}_{dj}\right]e^{-\mathrm{W}_d(\mathbf{Q})}\right|^2 \quad (2)$$

Where the sum on d is now a sum over the n_d atoms in a single molecule. If we now neglect interatomic correlations we obtain the following "incoherent approximation" to the coherent scattering cross section of a set of isolated molecules:

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k'}{k_0}\frac{N_m\sigma_c}{8\pi M}\frac{Q^2}{\omega}\left[n(\omega)+1\right]\sum_j(\omega-\omega_j)\sum_d\left|\hat{Q}.\hat{e}_{dj}\right|^2 e^{-2\mathrm{W}_d(\mathbf{Q})} \quad (3)$$

This further simplifies to read

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k'}{k_0}\frac{N_m n_d\sigma_c}{8\pi M}\frac{Q^2}{\omega}\left[n(\omega)+1\right]e^{-2\mathrm{W}}g(\omega) \quad (4)$$

where

$$g(\omega)=\frac{1}{3n_d}\sum_{\mu} m_\mu \delta(\omega-\omega_\mu) \tag{5}$$

is the normalized vibrational density of states; m_μ and ω_μ are, respectively, the degeneracy and the frequency of the μ th set of degenerate eigenmodes.

At energy transfers such that k' is much smaller than k_0, Q^2 is approximately proportional to ω. To the extent that the approximation represented by (4) can be justified, taking into account the $1/v$ cross section of the monitor counter, it follows that the intensity measured with a filter-analyzer spectrometer is approximately proportional to $\left[n(\omega)+1\right]e^{-2W}g(\omega)$. When $kT \gg \hbar\omega$, $n\omega \gg 1$, and the Debye-Waller factor changes slowly with Q. Then, if the scattering cross section may be reasonably represented by (4), the observed intensity is approximately proportional to its vibrational density of states.

The incoherent approximation is most easily justified when Q is large, and when the spectrometer averages data over a wide range of $\mathbf{Q}$. In filter-analyzer measurements the incoherent approximation applies best at high-energy transfers because in this case Q is large and the $\mathbf{Q}$–space volume sampled by the instrument is somewhat increased. At modest energy transfers studied [usually] it would not be surprising if coherent scattering effects produced noticeable changes in the intensities of peaks, but little change in peak positions is expected.

[1] B.N. Brockhouse. *In* Inelastic Scattering of Neutrons in Solids and Liquids. IAEA, Vienna 1961. pp. 113-151.

[2] A.D.B. Woods, B.N. Brockhouse, M. Sakamoto and R.N. Sinclair. *In* Inelastic Scattering of Neutrons in Solids and Liquids. IAEA, Vienna 1961. pp. 487-498.

[3] G.L. Squires. *Introduction to the Theory of Thermal Neutron Scattering*. Cambridge University Press, Cambridge. 1978. sect. 3.9

[4] S.W. Lovesey. *Theory of Neutron Scattering from Condensed Matter*. Clarendon Press, Oxford. 1984. Vol. 1, sect. 4.4